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Characterizing the Interfiber Bonding of Currency Paper Pulps

J. C. Smith and E. E. Toth

Polymer Science and Standards Division
Center for Materials Science
U.S. Department of Commerce
National Bureau of Standards
Washington, DC 20234

Progress Report Covering the Period
October 1, 1979 through September 30, 1980

November 1980

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Prepared for

Bureau of Engraving and Printing
U.S. Department of Treasury
Washington, DC 20401

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Table of Contents

	Page
Summary and Conclusions	i
Introduction	1
Materials Studied	2
Results From Scanning Electron Microscopy	4
Interaction of Water and Cellulose	5
Density of Bonding	7
Characterization of the Force-Elongation Curve	8
A Mechanism for Force-Elongation Behavior	11
Characteristic Lengths x_c for Low-Density Handsheets Made From Various Pulps	13
References	15

Summary and Conclusions

The nature of the adhesion between fibers in a currency pulp is different from that between fibers in kraft woodpulps. These differences are revealed by visual inspection using scanning electron microscopy, by studies of the effect of the beating process in the cellulose-water interaction in pulps and by the tensile behavior of low-density open-web handsheet specimens made from the various pulps.

A beaten currency stock was obtained, and pulp from this stock was fractionated by fiber length to obtain four fractions. Handsheets of 2.5 g/m^2 mass per unit area were prepared from the unfractionated stock, from each of the first four fractions and from a batch of stock from which fines passing through a 150 mesh screen were removed. Similar handsheets, for comparison purposes, were prepared from beaten and unbeaten Northern and Southern kraft woodpulps.

Morphologies of the handsheets and constituent pulp fibers were examined by scanning electron microscopy. These studies showed that the unfractionated currency pulp contained a large proportion of fines that collected at the fiber crossovers and appeared to act as a glue holding the fibers together. Fibers of the long-fiber-length fraction had thick cross-sections, but also were highly fibrillated, each fiber having many fine fibrils that formed bonds with adjacent fibers. This was in contrast to the morphologies of the woodpulps. Southern kraft pulp has coarse fibers that become ribbon-like after beating, but do not fibrillate. Bonding between Southern pulp fibers depends upon intimate contact between the fibers. Northern kraft pulp fibers have a finer texture and are more ribbon-like by nature, and do not require as much beating to induce flexibility and promote intimate interfiber contacts.

The morphological behavior of these pulps brought about by beating is in agreement with results obtained in a separate study of the cellulose-water interaction in the two types of pulps. During the preparation process pulp is subjected to mechanical action such as beating or refining. This treatment breaks down the structure of the fibers and permits them to imbibe water. In places where the crystalline microstructure is broken water will be strongly bound to the freshly formed crystalline surfaces, but where only the macrostructure is broken forming fissures or holes in the cell walls, the entering water is not strongly bound and may be regarded as free water. This free water is believed to cause swelling thereby imparting flexibility to the fiber and facilitating

interfiber conformability. It was found that beating causes increasing amounts of water to be imbibed by both cotton and woodpulp fibers. However in the cotton more of this additional imbibed water becomes bound water than is the case in woodpulp. This suggests that beating of woodpulp produces flexible conformable fibers, but causes fibrillation and fines formation in cotton fibers. Similar measurements have recently been carried out on microcrystalline cellulose, which simulates the fines produced by beating currency stock. Results are inconclusive but will be supplemented by additional research planned for next year.

Specimens 1 cm wide by 2 cm long from the handsheets were extended in a tensile tester. The force-elongation curves obtained had numerous force drops, each drop caused by the breakage of a bond between fibers constituting the handsheet network. The relative number of bonds per unit area ordinarily can be characterized by determining the average elongation between bond breaks. Measurements of this type were made without difficulty on specimens from the Southern pulp handsheets. However force-elongation curves for specimens from the currency pulp handsheets all had numerous force drops, mostly small and undifferentiated. It was not possible to obtain reliable representative measurements for these materials. This force-elongation behavior, though, is in accord with the behavior that might be expected of a highly fibrillated material held together by numerous small bonds.

Originally it was intended to characterize bond strength in terms of an energy parameter, obtained by averaging the energy dissipated by the fibrous network for each of a number of selected bond breaks, or by an alternative parameter obtained by averaging the force drops. These parameters could be useful for characterizing woodpulps, but would be difficult to determine for currency pulps. A more appropriate parameter might be related to the average force level at which bonds break. Work toward the development of this parameter is now in progress. As part of this work a considerable effort has been devoted to the problem of characterizing the force-elongation curve, up to the first bond break, of a fibrous network.

It has been found experimentally that when the slope F' of the force-elongation curve for a specimen from a low-density open-web handsheet is plotted versus the force F, the resultant plot can be closely fit by a straight line having the equation

$$\frac{dF}{dx} = \frac{1}{x_c}(C_2 + F)$$

where x is a distance variable measured in the direction of extension, and x_c and C_2 are constants. Thus, by integration, an equation for the force-elongation curve can be obtained in the form

$$F = C_2[e^{(x-x_o)/x_c} - 1]$$

where x_o is the value of the distance variable at which the force just increases from zero, so $x-x_o$ is the elongation of the network. The parameters x_c and C_2 are found by fitting a straight line to the plot of F' versus F, and the parameter x_o is found by fitting the equation of the curve (with x_c and C_2 already evaluated) to the force-elongation data.

If at intervals during a test on a handsheet specimen the extension is stopped, reversed until the tensile force in the specimen is zero, and then resumed, a series of force-elongation curves is obtained characterizing the specimen at various stages of deterioration during the test. It is found that x_o increases with each successive curve, indicating that the "unstrained" length increases during a test. x_c decreases slightly as the test proceeds, and C_2 decreases fairly rapidly.

x_c and C_2 are expected to be strongly dependent on the density or mass per unit area of the specimen. This was verified for x_c by measuring values of x_c for a series of specimens having densities varying between 1.25 and 2.50 g/m^2. x_c was found to decrease as the density increased. C_2 is expected to increase with density but this expectation has not been adequately checked experimentally. If x_c and C_2 are sensitive to specimen density changes they should vary from specimen to specimen because of fluctuations in the local density of the handsheet from which the specimens are taken. This suggests the possibility of using x_c and C_2 to reduce adhesion parameters determined on different specimens to a standard value, and thus improve the precision of measurements on a given handsheet.

The parameter x_c has the dimensions of length and can be referred to as a characteristic length. This characteristic length appears to increase or decrease with the average size of the meshes in a specimen fiber network. For instance, x_c was measured on specimens from Northern and Southern kraft pulp handsheets, both of 2.5 g/m^2 density. x_c for the Southern pulp was larger than the x_c for Northern pulp. This is because fibers of Southern pulp are coarser than fibers of Northern pulp; hence a Southern pulp handsheet has fewer fibers per unit area and larger

meshes than does a Northern pulp handsheet. In another instance it was shown by measurements of x_c that beating decreases the mesh size of Northern and southern pulp handsheets, presumably by increasing the proportion of fiber crossovers that are bonded. Measurements of x_c on handsheets from currency pulp fractions showed that the mesh size of the handsheets made from fraction III (short fibers) was appreciably less than that for the others. x_c appears to be an especially useful and easily measured parameter for use in these studies, and further applications of it will be investigated.

Introduction

Paper used to make currency should have a good balance between durability and cost, as the replacement of worn out currency is an expensive operation. Therefore it would be desirable to identify and characterize those aspects of the morphology of paper networks that affect the durability and other desirable properties, and to obtain a better understanding of how these properties are controlled by the manufacturing process. Researches of this nature might contribute to the development of durable paper made from blended woodpulps instead of the more expensive cotton and linen pulps presently used.

During the past year these researches have concentrated on studying the properties of very thin low-density open-web handsheets made from currency pulp and from Northern and Southern kraft woodpulps. Samples from the handsheets were extended in a tensile tester to obtain force-elongation curves from which data on interfiber adhesion could be obtained. Mathematical relationships describing force-elongation behavior were derived, and a parallel-spring model for elucidating this behavior was proposed. Preliminary values were obtained for some parameters of use in characterizing interfiber bonding. Morphologies of the handsheets and constituent fibers were examined by scanning electron microscopy. Some research on the cellulose-water interaction was also carried out. This progress of research conducted this year is reported in the following text.

Materials Studied

Currency stock was obtained from Crane and Co., Inc. The stock was collected after the beater, but before passing through the refiners and jordan. A portion of this stock was fractionated in a Bauer-McNett classifier using screens of mesh sizes 14, 35, 65 and 150. Four fractions were obtained. A fifth fraction consisting of fines passing through the 150 mesh screen was not collected. Classification data are given in table 1. Open-web handsheets were prepared from the unfractionated material, from each of the first three fractions, and from a pulp obtained by a separate classification in which only the fines were removed.

Two types of woodpulp, Northern and Southern softwood kraft pulps, were also used in these studies. Batches of these pulps at a consistency of ten percent were subjected to beating in a PFI laboratory beater for the desired number of revolutions. There was no clearance between the bedplate and roll, force was 33 N (3.4 kg), and the relative velocity of roll to bedplate was 6 m/s.

Handsheets were formed from the woodpulps as follows: A 33 x 33 cm sheet of filter paper (Whatman No. 541) was placed on top of the support wire of the deckle box and wetted. The deckle box was closed, clamping the sheet firmly at the bottom. Water was then added to the box. An appropriate amount of pulp was placed in approximately one liter of water and disintegrated in a British disintegrator for 7500 revolutions for beaten pulp, or 25,000 revolutions for unbeaten pulp. Whenever unbeaten pulp was used it was first soaked overnight in a liter of water prior to disintegration. The disintegrated pulp was added to the water in the deckle box, agitated carefully, and then drained through the filter paper.

The filter paper and deposited pulp fibers were placed on a cushion of pulpsheet. A 33 x 33 cm ferrotype plate was placed carefully over the top of the filter paper and layer of pulp fibers. Another pulpsheet cushion was placed over the top of the ferrotype plate, and the entire sandwich was placed in a hydraulic press for five minutes at a pressure usually near 0.34 MPa (50 lb/in^2). During the pressing the layer of fibers transferred to the ferrotype plate. It was allowed to dry and was then carefully removed. In some cases the handsheet did not transfer to the ferrotype plate, but pretreating the filter paper with a fluorocarbon spray lubricant usually eliminated this difficulty.

Densities of most of the handsheets manufactured in this way were 2.5 g/m^2 mass per unit area, but for some special purposes other handsheets of densities

varying between 1.0 and 5.0 g/m^2 were also prepared. Handsheets in this density range have open webs, and are only two or three fibers thick in most places.

The process just described was used to prepare handsheets from the unfractionated currency pulp, from each of the first three fractions and from a reconstituted pulp made from the first four fractions recombined in their original proportions. It was more difficult to prepare these handsheets because the webs did not transfer readily to the ferrotype plate, and those that did transfer could not be easily removed subsequently. This was particularly true of the webs made from the unfractionated pulp. However it was found that if the filter paper was given a heavy coating of Scotchgard* transfer was achieved, and sufficiently large pieces of handsheet could later be removed from the ferrotype plate.

* Proprietary water repellant containing fluoroaliphatic resin, manufactured by 3 M company.

Results From Scanning Electron Microscopy

Specimens from the currency pulp handsheets were photographed in a scanning electron microscope. For comparison purposes SEM photographs were also obtained for similar handsheet specimens of Southern kraft pulp, unbeaten and beaten 5000 revolutions in a laboratory beater. Figure 1 is a micrograph of an unfractionated currency pulp specimen, and figure 2 is a micrograph of a specimen reconstituted from the first four fractions. Most of the fines material in figure 1 is not present in figure 2. The appearance of these micrographs suggests that the fines act as a glue filling the interstices between pulp fibers and helping cement them together. Figure 2 shows that the pulp fibers have rounded cross-sections that do not always make intimate contact, but that fibrillation provides many minor adhesions between fibers. The unbeaten Southern pulp specimen is shown in figure 3. The fibers in this specimen have a rounded cross-section and do not make intimate contact, so that fiber cross-overs are not strongly bonded. The effect of beating the Southern pulp is shown in figure 4. The fibers here have been flattened into ribbons that make better contact and provide stronger bonds, but the beating has not caused fibrillation, and the frequent minor adhesions shown in figure 2 are not present. The appearance of Northern pulp fibers is similar except that the unbeaten fibers are flatter, and subsequent beating enhances this ribbon-like structure.

It should be recalled that when beaten Southern pulp handsheets were prepared, the webs transferred readily to the ferrotype plate, and were easily removed subsequently. When handsheets were prepared from unfractionated currency pulp the webs would not transfer unless the filter paper bed had previously been heavily coated with a water repellent. This reinforces the idea that bonding in currency pulps is facilitated by the hydrophylic fines acting as a glue, and is also enhanced by a large number of bonds to the thin fibrils produced by a beating treatment. Thus the bonding mechanism in currency paper is different from that in wood pulps where bond strength depends on intimate contacts between pliable ribbon-like fibers relatively free of small fibrils.

Interaction of Water and Cellulose

During the preparation process, pulp is subjected to mechanical action such as beating or refining. This treatment breaks down the structure of the fibers and permits them to imbibe water. In places where the crystalline microstructure of the fiber is broken water will be strongly bound to the freshly formed crystalline surfaces, but where only the macrostructure is broken to form fissures or holes in the fiber cell walls the entering water is not strongly bound and may be regarded as free water. This free water is believed to cause swelling thereby imparting flexibility to the fibers and facilitating interfiber conformability. The way in which different pulps respond to a beating treatment can be studied by measuring the proportions of bound and free water in different pulp samples.

The water imbibed by a pulp specimen in the swollen state is measured by solute exclusion. A pulp specimen is added to a solution of dextran in water, and the excess water in the specimen dilutes the solution. The amount of dilution is measured by polarimetry, and the imbibed water, or fiber saturation point, of the specimen is calculated. Part of this imbibed water is tightly bound to the cellulose and does not freeze at temperatures down to 233 K. This bound water fraction is measured by differential scanning calorimetry. The difference between the fiber saturation point and bound water is considered to be free water.

In some previously reported work [1] it was found that beating causes increasing amounts of water to be imbibed by both cotton and woodpulp fibers. However beating causes an increasing fraction of the water imbibed to be bound in cotton. This does not happen to as great an extent in woodpulp. Thus beating of woodpulp produces flexible conformable woodpulp fibers, but causes fibrillation and fines formation in cotton fibers.

Some more recent work has been concerned with achieving a better understanding of what the fiber saturation point of a sample actually measures. In determining the total amount of water within pulp fibers in the swollen state (fiber saturation point) it is assumed that water bound to the surface of the fibers and in contact with the dextran solution behaves as free water, and will take part in the dilution of the dextran solution. If water bound to the surface does not dilute the dextran solution the calculated value for the fiber saturation point will be misleading. The magnitude of the discrepancy involved will depend upon the extent of the surface area.

In an effort to determine whether water bound to the surface does dilute the dextran solution, bound water and the fiber saturation point were determined on a commercial sample of microcrystalline cellulose. Presumably the crystalline cellulose is nonporous, and no water would be expected to enter the crystalline lattice. If water bound to the surface of the crystalline cellulose does not enter into the dilution of the dextran solution, the bound water content and the fiber saturation point would by nominally equal. If the surface bound water does dilute the dextran solution there would be no fiber saturation point.

For the commercial sample of microcrystalline cellulose the bound water content was 0.25 g H_2O/g cellulose, while the fiber saturation point was 0.56 g H_2O/g cellulose. These results indicate that not only the bound water but additional water, such as perhaps loosely bound water, does not enter into the dilution of the dextran solution, or that the microcrystalline cellulose is porous and is penetrated by water.

Density of Bonding

Specimens from the handsheets were tested in a tensile tester to obtain recordings of force versus elongation. Figure 5 shows the result of a test run on a 2-cm specimen of unfractionated currency pulp. Full scale force is 49 mN (5 g), and 1 cm of chart travel corresponds to a specimen elongation of 0.01 cm. The numerous "jags" or force drops indicated on the curve correspond to interfiber bond breaks; thus the average elongation between breaks should provide a measure of the density of bonding per unit area of the test specimen. In this case there are frequent jags of small magnitude suggesting the presence of numerous minor adhesions between the constituent fibers. Force-elongation recordings for specimens from the other currency pulp handsheets were similar.

Figure 6 shows the result of a test run on a 2-cm specimen of Southern kraft pulp beaten 5000 revolutions in a laboratory beater. The jags in this case are not so frequent or so small in magnitude, suggesting the relative absence of minor adhesions.

For handsheets of the same mass per unit area the density of bonding per unit area should depend upon the coarseness (mass per unit length) of the fibers forming a web, and may also be slightly dependent upon the average length of the fibers. In addition, the average elongation between bonds, which gives an indication of bonding density, should be larger for narrow specimens than for wide ones. The average elongation per break was measured on the currency pulp specimens to see if differences in the density of bonding could be detected. The results of these measurements are given in table 2. No obvious differences in density of bonding are revealed by these measurements. However it should be noted that the values given for specimens of different width are essentially the same. A possible interpretation of this is that the numerous breaks due to minor adhesions are swamping the data. Under these conditions it will be necessary to devise a consistent means of screening out the jags due to minor adhesions in order to obtain more meaningful results.

Measurements on specimens from the Southern kraft pulp handsheets are given in table 3. These results show that beating greatly increases the number of bonds at fiber cross-overs. Also, the elongation per break is seen to increase as the specimen width decreases, in accordance with expectations but in contrast with the behavior observed in the tests on currency pulps. This difference in behavior may be attributed to the fact that beating Southern pulp fiber does not produce the kind of fibrillation observed in the beaten currency pulp.

Characterization of the Force-Elongation curve

A previous report [2] has discussed various bonding parameters that can be derived from tensile test data. The bond strength could be characterized in terms of an energy parameter, obtained by averaging the energy dissipated by the fibrous network for each of a number of selected bond breaks, or in an alternative characterization the drops in force could be averaged. An examination of figure 5 however shows that the force drops resulting from breakage of bonds between the pulp fibers are obscured by the many small drops resulting from bond breaks in which tiny fibrils are involved. For this reason a different bonding parameter such as the force level at which the principal bonds break might be more appropriate. In order to develop this and other parameters and to increase our knowledge of the mechanical behavior of fiber networks, much of this year's research effort was devoted to the problem of characterizing the force-elongation curve of a fibrous network.

Consider the force-elongation curve, figure 7, obtained by testing a specimen from a 2.5 g/m^2 handsheet of Northern kraft pulp. The specimen was 2 cm long and 1 cm wide. 1 cm of chart travel corresponds to a specimen extension of 0.01 cm. Full scale force for the specimen was 98 mN (10 g). At intervals the test was stopped, the extension was reversed until the tensile force in the specimen was zero, and the specimen then reextended to obtain a new force-elongation curve. Recording traces during reversals of the cross-head travel are not shown.

The general condition of the specimen at various stages is noted on the figure. For the first five reextensions the specimen did not develop any large holes or tears, and the state of the specimen at the beginning of the reextension is described as "intact". At later stages of the test the specimen developed holes, and tore at the edges until in the final stages the tensile force was sustained by only a few parallel fiber paths collapsed together into filaments.

In figure 8 are shown plots of slope F', or force per unit elongation, versus force F for each of the reextended force-elongation curves of figure 1. The origin of the F' scale has been shifted vertically for each of these plots, and arbitrary units have been selected to display the plots to best advantage. The curve corresponding to each plot is identified by its region of chart travel. The plots are seen to be linear and to have positive intercepts on the F' axis. Data from the initial force-elongation curve obtained before any reextension does not plot satisfactorily and is not shown.

Similar behavior has been observed in tests on handsheets of various masses per unit area formed from the different pulps. The linearity of the plots obtained from these tests is not affected by the amount of deterioration incurred by the specimen during extension. In many instances the slopes corresponding to initial and final forces do not plot on a straight line, but the intermediate data are linear to a good approximation. The intercepts on the F' axis are usually positive, and tend toward zero as the specimen deteriorates. In a few instances the intercepts become negative. This latter condition is probably an artifact. It can be caused, for instance, by a specimen with a large tear extending inward from one side. Such a specimen requires a large initial extension before the network structure becomes appreciably strained.

It has been found from many tests that F', F data from the initial force-elongation curve in a series often does not plot on a straight line. In some instances a straight-line plot is obtained but the slope of the plot is much steeper than the slopes of F', F plots for subsequent reextension curves. It is only after the specimen has been "broken in" that data can be obtained from subsequent curves. In the test of figure 7 the first reextended curve was obtained before the specimen was completely broken in, and the curve is similar to one that would be obtained by an additional force acting in parallel with the tensile forces sustained by the specimen network. This strong parallel force may involve bending forces and other forces required for an initial alining and shaping of the specimen.

Force-elongation curves of these paper network specimens exhibit visco-elastic behavior. Thus the shapes of the reextension curves will depend on the waiting time between halting of an extension and the starting of reextension. In order that experimental results be as consistent as possible the waiting times should be approximately the same for all reextension curves. The effects of visco-elasticity are not considered in the theory presented here, but their possible influence upon experimental results should be kept in mind.

Let the equation of a straight line faired through a plot such as those shown in figure 8 be

$$\frac{dF}{dx} = \frac{1}{x_c}(C_2 + F) \qquad (1)$$

where x is a distance variable measured in the direction of extension. x_c is the reciprocal of the slope of the straight line, and C_2/x_c is the intercept on the

F' axis. Eq (1) has the solution

$$F = C_1 e^{x/x_c} - C_2 \qquad (2)$$

where C_1 is a constant to be evaluated from data such as figure 7.

Table 4 illustrates how data from the curve between 7.0 and 13.0 cm chart reading on figure 7 can be fitted by a curve of the type, eq (2). The first column lists cross-head displacements, and column 2 lists corresponding values of force obtained from the force-elongation curve. Intermediate values of force obtained by interpolation are listed in column 3. The slopes in column 4 are obtained by dividing successive force differences by differences in cross-head displacement. The data of columns 3 and 4 are plotted, as in figure 8, and a least-squares fit of eq (1) yields the values: $C_2/x_c = 350.48$ mN/cm, $x_c = 0.033434$ cm. Eq (2) can then be fit by the least-squares technique to data in columns 1 and 2 to yield the value $C_1 = 1.0636$ mN. The fitted equation is used to calculate the forces in column 5. In this instance a good fit is apparent.

According to eq (2), F becomes zero when

$$x = x_o = x_c \ln(C_2/C_1) \qquad (3)$$

Thus eq (2) can be put in the form

$$F = C_2[e^{(x-x_o)/x_c} - 1] \qquad (4)$$

x_o is the value of the distance variable at which the force just increases from zero, so $x - x_o$ is the elongation of the network. As the specimen fibers become better alined, and as the specimen deteriorates with increasing stretch, the unstrained length of the specimen increases. Thus for each successive reextension curve of figure 7, successively increasing values of x_o should be expected.

Ideally the successive values of x_o could be calculated using eq (3). Table 5 provides an example. Here are tabulated the values of C_1, C_2, x_c and x_o calculated from each of the reextension curves of figure 7. The agreement between values of x_o in cm of chart with the apparent values obtained by visual inspection of figure 7 is generally good. However the value of x_o calculated for the curves in the 27.0-35.0 cm range is too low. In this case the curve, eq (2), that best fits the data over most of the range does not fit well initially. This is probably an effect due to detericration of the specimen.

A Mechanism for Force-Elongation Behavior

When a fibrous network is stretched segments of the fibers composing the network tend to orient themselves in the direction of the extension, and to resist the extension through the combined action of forces along their lengths. As the extension proceeds more and more of these segments between bonds become oriented and bear load. As a result of this process the force-elongation curve has a slope that increases with increasing elongation. Thus the reaction of the network to extension could be modeled by a system of parallel filaments of unequal length, each filament adding to the resistive force of the system as the extension is increased. The stiffness and the degree of orientation of the network fibers also contribute to the network's reaction to stretching, but these and other effects will be considered as secondary in importance and usually be disregarded.

Consider a system of parallel filaments. Let the length of the shortest filament, or unstrained length of the system be l_o, and let the length of any other filament be $l = l_o + \eta$. Let the number of filaments having lengths between l and $l + d\eta$ be $f(\eta)d\eta$. Let the filaments all have the same spring constant k, neglecting the decrease in k with increase in unstrained length. If this system of parallel filaments is extended from a length l_o to a length $l_o + \xi$ the sum S of the tensile forces in the filaments is

$$S = \int_o^{\xi} kf(\eta)(\xi - \eta)d\eta \tag{5}$$

If it is assumed that for $\eta > 0$

$$f(\eta) = (C_2/kx_c^2)e^{\eta/x_c} \tag{6}$$

eq (5) becomes

$$S = (C_2/x_c^2)\int_o^{\xi} e^{\eta/x_c}(\xi - \eta)d\eta = C_2[e^{\xi/x_c} - 1] - C_2(\xi/x_c)$$

or $$(C_2/x_c^2)\int_o^{\xi} e^{\eta/x_c}(\xi - \eta)d\eta + C_2(\xi/x_c) = C_2[e^{\xi/x_c} - 1] \tag{7}$$

If ξ is set equal to $x - x_o$ the expression on the right hand side of this equation is the same as the expression, eq (4), for the force in a fibrous network. This can be interpreted to mean that the force in a fibrous network can be simulated by a system of parallel filaments each of spring constant k and having a length distribution function given by eq (6), acting in parallel with a spring of spring constant C_2/x_c and unstrained length l_o.

This spring may be considered as composed of n filaments each of length ℓ_o and spring constant k. That is,

$$C_2/x_c = nk \tag{8}$$

Thus when a fibrous network specimen is elongated by an amount ξ, the number of extended filaments in the model representing the network is

$$N = n[1 + \frac{1}{x_c}\int_0^{\xi} e^{\eta/x_c} d\eta] = ne^{\xi/x_c} = \frac{C_2}{kx_c} e^{\xi/x_c} \tag{9}$$

The values of the parameters C_2 and x_c are dependent upon the density of distribution of the parallel filaments modeling the handsheet test specimen. Suppose, for instance, that there are two specimens A and B taken from two handsheets of different masses per unit area made from the same pulp furnish. Let specimen B have the greater density. The parameters x_{cB} and C_{2B} might be expected to have the following relationships to the corresponding parameters for specimen A:

$$x_{cB} = \frac{1}{\alpha} x_{cA} \tag{10}$$

$$C_{2B} = \beta C_{2A} \tag{11}$$

where α and β are functions of the density ratio D_B/D_A and have values greater than unity.

According to the assumptions, eqs (10) and (11), the distribution functions, eq (6), for the two specimen models are related as

$$F_B(\eta) = \frac{C_{2B}}{kx_{cB}^2} e^{\eta/x_{cB}} = \alpha^2\beta \frac{C_{2A}}{kx_{cA}^2} e^{\eta\alpha/x_{cA}} \tag{12}$$

Thus to obtain the distribution function for the specimen B from that for specimen A, the scaling factor x_{cA} should be multiplied by $1/\alpha$, and the coefficient C_{2A} by β. This effectively raises the exponential term by the factor α and the constant multiplier by the factor $\alpha^2\beta$. The scaling factor has the dimensions of length, and will sometimes be refered to as a "characteristic" length.

Characteristic Lengths x_c for Low-Density Handsheets Made From Various Pulps

Characteristic lengths x_c were obtained by measuring the slopes of the F' versus F plots for a number of test specimens. The value of x_c for a given specimen was taken as the average of the x_c values for the first few reextension curves. Only reextension curves taken when the specimen was "intact" were used. The value of x_c tended to decrease slightly for each of the successive reextension curves, as may be seen in the data of table 5, but this was not believed to affect significantly the average values of x_c that were obtained. Table 6 lists the characteristic lengths obtained from tests on a series of Northern kraft pulp handsheets of densities from 1.25 to 2.50 g/m^2. According to eq (10) values of x_c should decrease as the density of the handsheet increases. The experimental values are in agreement with eq (10) for densities of 2.0 g/m^2 or less. This is more easily seen in the plot of x_c versus handsheet density, figure 9. More tests are needed to check eq (10) at densities greater than 2.5 g/m^2.

Tables 7 and 8 list characteristic lengths x_c for Northern and Southern kraft pulp handsheets of 2.5 g/m^2 density made from pulps subjected to different amounts of beating. The Northern pulp handsheets made from pulp beaten 1000, 2000 and 5000 revolutions in a laboratory beater had approximately the same values for x_c, but the characteristic length for the handsheet made from pulp beaten 10,000 revolutions was slightly lower in value. Although all the handsheets had the same density, or mass per unit area, more fiber cross-overs were bonded in the handsheet made from the highly beaten pulp, resulting in a network with a shorter average mesh length, and smaller values of x_c for the specimens tested. The results for Southern pulp handsheets were similar, as may be seen from table 8.

According to tables 7 and 8 the characteristic length x_c for a 2.5 g/m^2 handsheet specimen of Northern pulp is appreciably less than the value of x_c obtained for a 2.5 g/m^2 specimen from a handsheet of Southern pulp. Southern pulp is coarser than Northern pulp; hence a Southern pulp handsheet of given mass per unit area will consist of thicker but fewer fibers, and its average mesh length and the associated characteristic length for the specimen will be larger.

Table 9 gives the characteristic lengths x_c for handsheet specimens made from the currency pulp fractions listed in table 1. The values of x_c vary from handsheet to handsheet in a way that might have been expected, but the precision of measurement is usually too low to interpret these variations with much

certainty. The characteristic length for the fraction III handsheet is noticeably lower than that for any of the others. The average length of the fibers in this fraction is very short; thus there are a large number of fibers per unit mass which might be expected to form a network of small average mesh size.

With less certainty one is tempted to note that x_c for the unfractionated handsheet is the same as that for the handsheet made from fractions I, II, III and IV. Fines is a significant part of the mass in the handsheet of unfractionated pulp, so one might expect the fibers to form a network of larger mesh size than in the handsheet formed from the same fibers but no fines. Possibly the fines improve the bonding so that fiber cross-overs in the sheet of unfractionated pulp are more efficiently bonded. One might note also that the characteristic length x_c is larger in the handsheet made from fraction I than in the handsheet made from fractions I, II, III and IV. The fibers of fraction I are longer so there are fewer fibers per unit mass, and these fibers might form a network with a larger average mesh size.

References

1. E. L. Graminski and E. E. Toth, The Cellulose-Water Interaction in Rag and Wood Pulps, NBSIR 80 - 1970.

2. J. C. Smith, Characterizing the Interfiber Bonding of Paper Pulps: Rationale for Bonding Parameters derived From Tensile Test Data, NBSIR 79 - 1722.

Table 1.

Classification of Crane Currency Stock

	Weight of Wet Pulp g	Moisture Content %	Weight of Dry Pulp g
Unfractionated pulp	924.	76.4	218.
Fraction I (#14 mesh)	365.	78.3	79.
Fraction II (#35 mesh)	260.	78.9	55.
Fraction III (#65 mesh)	94.	74.5	24.
Fraction IV (#150 mesh)	45.	73.1	12.
Fines			48.

Table 2.

Elongation per Break for Currency Pulp Handsheet Specimens of Various Widths [1/]

Description and Sample Number		Elongation per Break ± Standard Deviation		
		1 cm width %/break	0.5 cm width %/break	0.3 cm width %/break
Unfractionated	1.	0.099±0.009	0.098±0.011	0.115±0.019
	2.	0.098±0.015		
Fraction I	1.	0.122±0.016	0.115±0.024	0.123±0.012
	2.	0.092±0.011		
Fraction II	1.	0.110±0.007	0.092±0.011	0.108±0.014
	2.	0.096±0.012		
Fraction III	1.	0.083±0.007	0.087±0.009	0.086±0.011
Fractions I, II, III, IV Recombined	1.	0.099±0.016	0.097±0.004	0.086±0.019
	2.	0.096±0.013		

1. Values given are the averages of determinations from tests on 5 different specimens.

Table 3.

Elongation per Break for Southern Kraft Pulp Handsheet Specimens of Various Widths [1/]

Description	Elongation per Break ± Standard Deviation		
	1 cm width %/break	0.5 cm width %/break	0.3 cm width %/break
Unbeaten	1.342±0.544		
Beaten [2/]	0.187±0.028	0.245±0.036	0.399±0.132

1. Values given are the averages of determinations from tests on 5 different specimens.
2. Pulp was beaten 5000 revolutions in a laboratory beater.

Table 4.

Fit of Curve Between 7 Cm and 13 Cm on the Chart, Figure 7.

X-Head Distance x cm	Force F mN	Interp. Force F mN	Slope F' mN/cm	Fitted Force F mN
.082	4.9			4.95
.084		5.93	515.	
.086	7.0			7.06
.088		8.19	613.	
.090	9.4			9.45
.092		10.79	686.	
.094	12.2			12.14
.096		13.68	760.	
.098	15.2			15.18
.100		16.92	858.	
.102	18.6			18.59
.104		20.59	981.	
.106	22.6			22.45
.108		24.81	1128.	
.110	27.1			26.79
.112		29.37	1152.	
.114	31.7			31.68
.116		34.47	1397.	
.118	37.3			37.20
.120		40.31	1520.	
.122	43.3			43.41
.124		46.78	1716.	
.126	50.2			50.42
.128		54.28	2035.	
.130	58.4			58.32

Least-squares fit to data in columns 3 and 4: $F' = 350.48 + F/.033434$

Least-squares fit to data in columns 1 and 2: $F = 1.0636 \exp(x/.033434) - 11.718$

Table 5.

Values of C_1, C_2, x_c, x_o for the Reextension Curves, Figure 7

Curve Chart Position cm	C_1 mN	C_2 mN	x_c cm	Cross-Head x_o cm	Chart x_o cm
3 - 8	1.8405×10	40.193	.046814	.036566	3.7
7 - 13	1.4344	11.718	.033434	.070225	7.0
11 - 18	3.8282×10^{-1}	12.007	.033305	.11476	11.5
15 - 23	2.7726×10^{-2}	6.0772	.029821	.16073	16.1
22 - 29	1.7457×10^{-3}	3.8832	.028384	.21876	21.9
27 - 35	2.3254×10^{-5}	.32557	.025924	.24749	24.8
34 - 42	1.6423×10^{-5}	.93938	.030272	.33160	33.2
40 - 47	2.4548×10^{-5}	1.5966	.035909	.39797	39.8

Table 6.

Characteristic Length x_c for Northern Kraft Pulp Handsheets of Different Densities

Density g/m^2	Characteristic Length, x_c cm
1.25	0.048 ± 0.007
1.50	0.042 ± 0.006
1.75	0.034 ± 0.006
2.00	0.029 ± 0.004
2.50[1]	0.029 ± 0.003[2]
2.50	0.029 ± 0.004

Unless otherwise noted handsheets were prepared from pulp beaten 5000 revolutions in a laboratory beater, and the mats were pressed 5 minutes at 350 kPa. Specimens were 2 cm long and 1 cm wide. Values given are the averages and standard deviations from measurements on 5 specimens.

1. Pressed at 44 kPa.
2. Average of 9 specimens.

Table 7.

Characteristic Length x_c for Northern Kraft Pulp Handsheets Made From Beaten Pulps

Amount of Beating	Characteristic Length, x_c cm
1,000 rev	0.035 ± 0.005
2,000 rev	0.030 ± 0.005
5,000 rev	0.029 ± 0.003[1/]
5,000 rev[2/]	0.029 ± 0.004
10,000 rev[3/]	0.023 ± 0.002

Table 8.

Characteristic Length x_c for Southern Kraft Pulp Handsheets Made From Beaten Pulps

Amount of Beating	Characteristic Length, x_c cm
1,000 rev	0.043 ± 0.008
2,000 rev	0.043 ± 0.007
5,000 rev	0.040 ± 0.004
10,000 rev[4/]	0.032 ± 0.005

Unless otherwise noted handsheets of 2.5 g/m^2 density were made from pulp beaten the stated number of revolutions in a laboratory beater. Mats were pressed 5 minutes at 44 kPa. Values given are the averages and standard deviations on 5 specimens 2 cm long and 1 cm wide.

1. Average of 9 specimens.
2. Pressed 5 min at 350 kPa.
3. Pressure not known.
4. Mat laid on cotton linters and pressed 5 min at 660 kPa.

Table 9.

Characteristic Length x_c for Handsheets
Made From Different Currency Pulp Fractions.

Fraction	Characteristic Length, x_c cm
Unfractionated	0.020 ± 0.003[1/]
Fraction I	0.027 ± 0.006
Fraction II	0.024 ± 0.003
Fraction III	0.014 ± 0.003
I, II, III, IV recombined	0.021 ± 0.003

Handsheets were of density 2.5 g/m^2. Mats were pressed 5 minutes at 350 kPa. Values given are the averages and standard deviations from measurements on 5 specimens 2 cm long and 1 cm wide.

1. Average of 6 specimens.

Figure 1.
Currency Pulp, Unfractionated
195 X

Figure 2.
Currency Pulp,
Recombined Fractions, I, II, III, IV
180 X

Figure 3.
Southern Kraft Pulp, Unbeaten
185 X

Figure 4.
Southern Kraft Pulp,
Beaten 5000 Revolutions
195 X

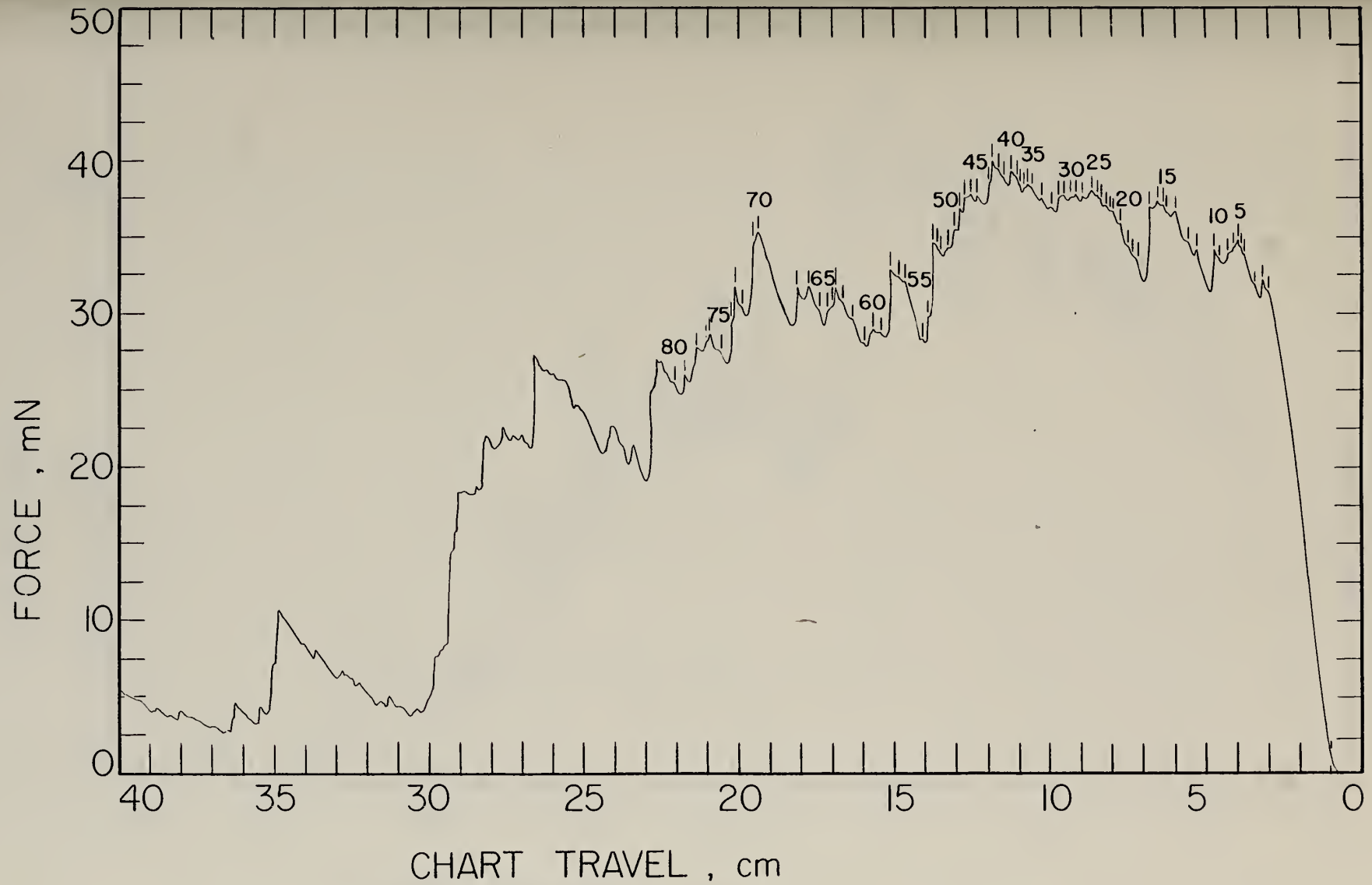

Figure 5. Force-elongation curve for a handsheet specimen of unfractionated currency pulp, 2.5 g/m^2 mass per unit area.

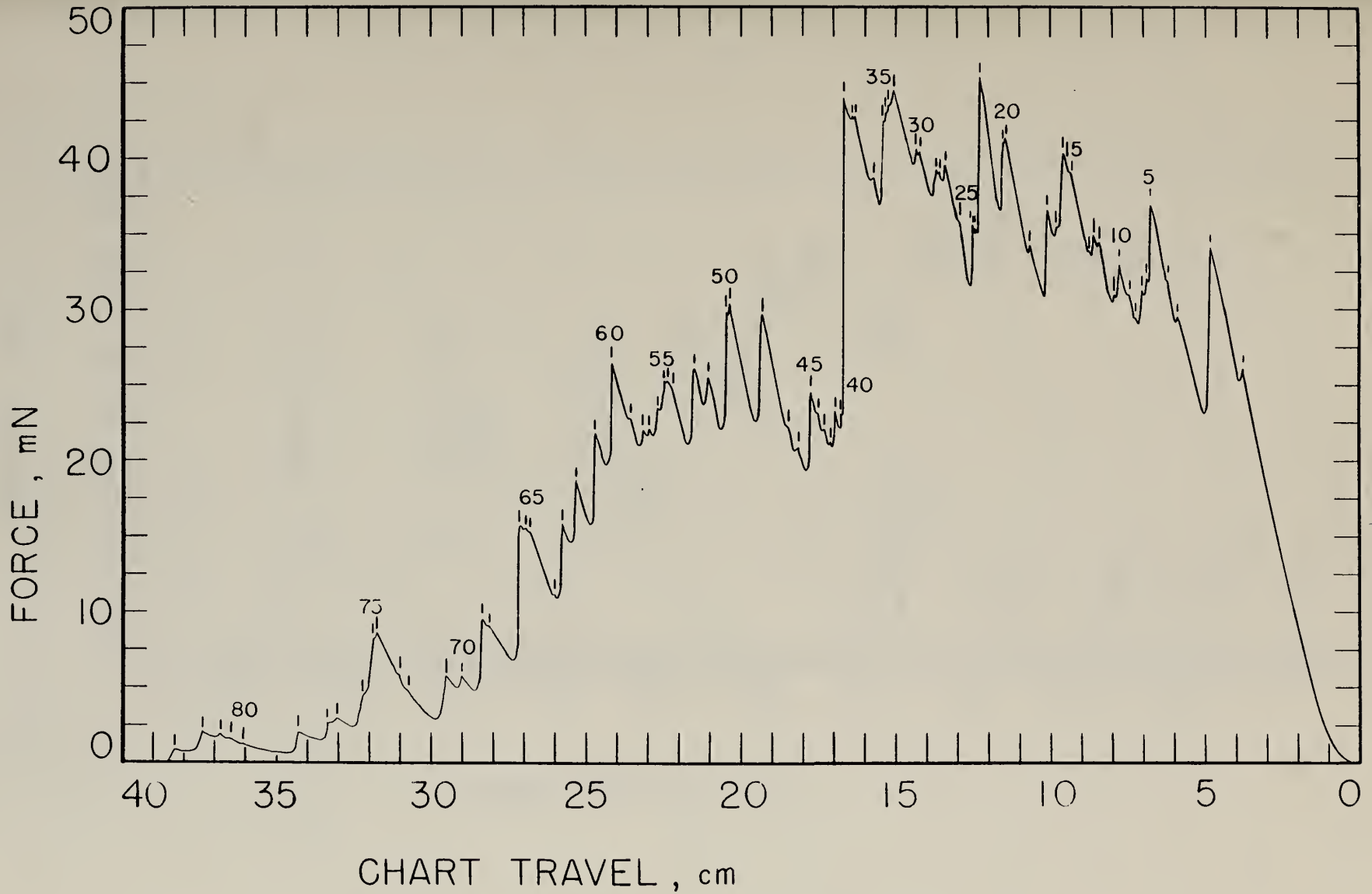

Figure 6. Force-elongation curve for a handsheet specimen of beaten Southern kraft pulp, 2.5 g/m^2 mass per unit area.

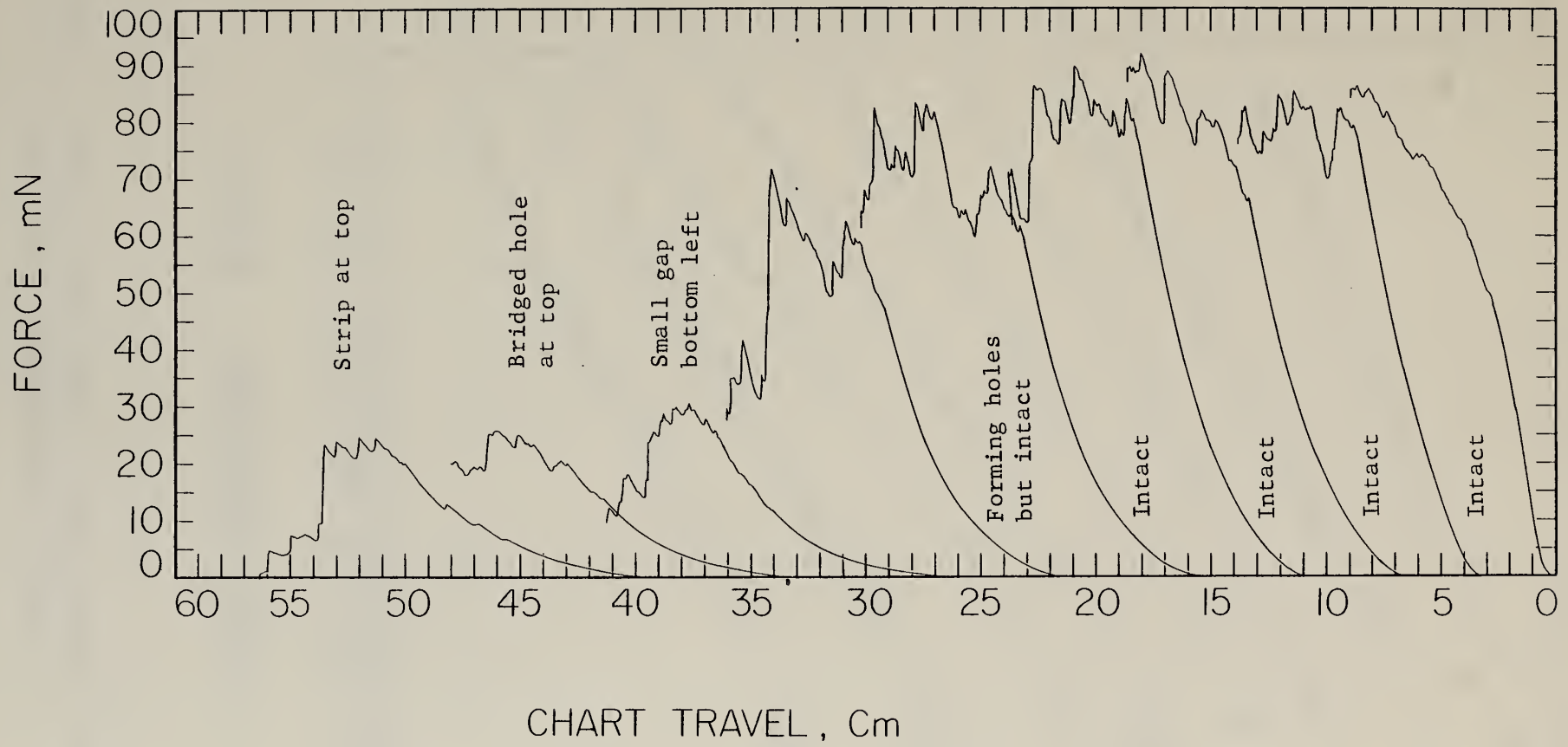

Figure 7. Force-elongation curve for a handsheet specimen of Northern kraft pulp, 2.5 g/m^2 mass per unit area.

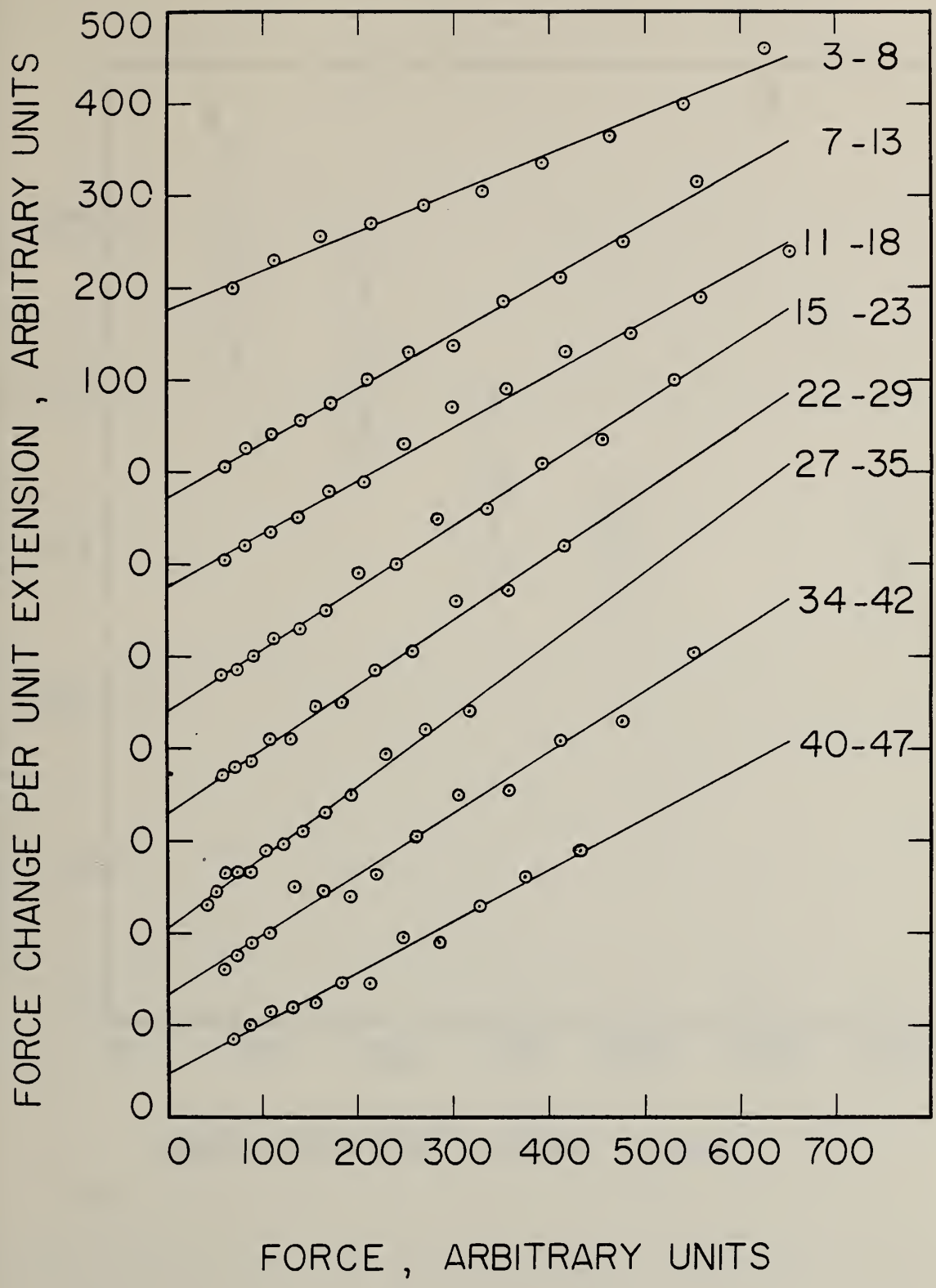

Figure 8. Plots of force change per unit extension versus force for reextension curves, figure 7.

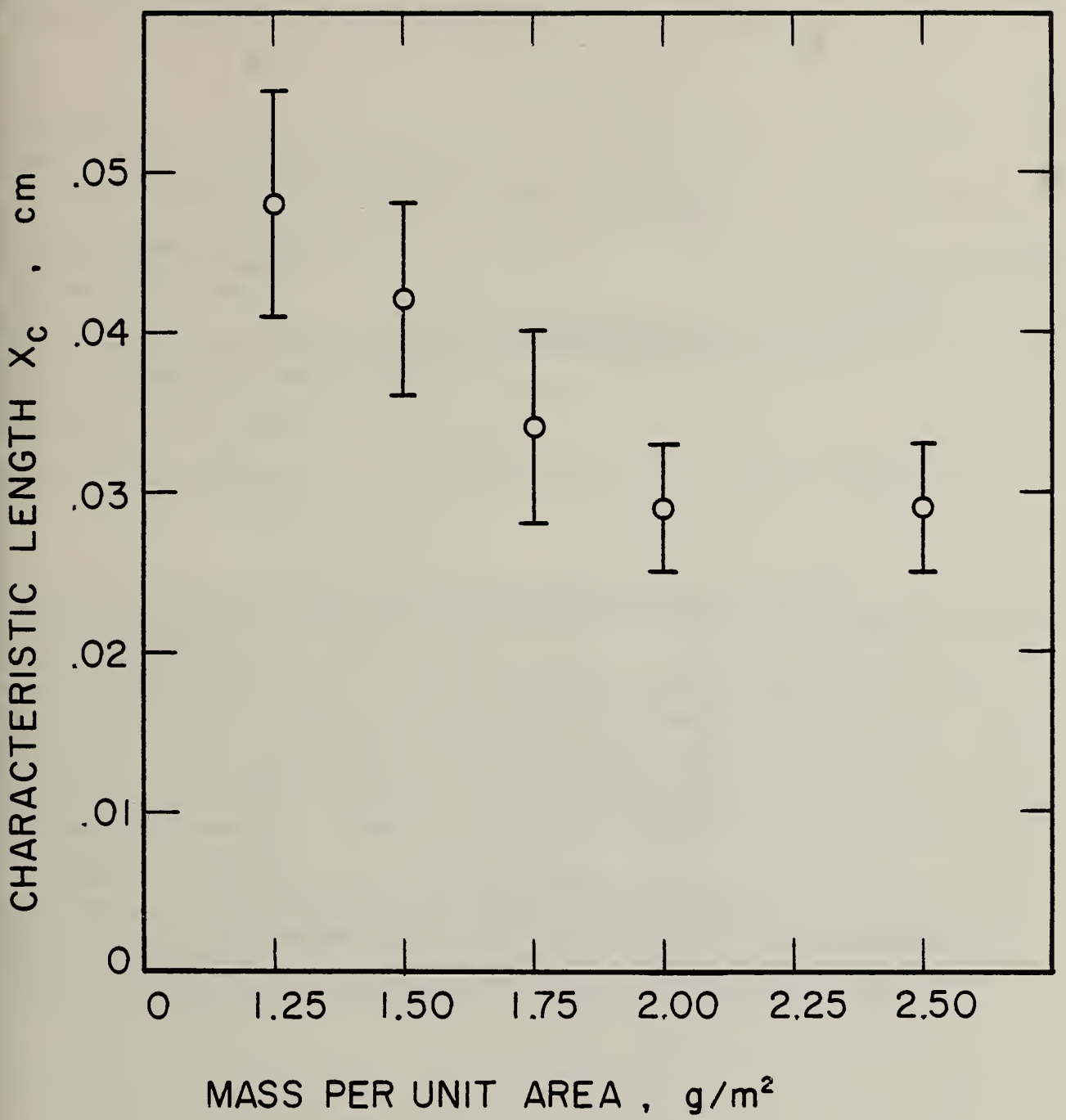

Figure 9. Plot of characteristic lengths x_c for handsheets of different densities, from table 6.

NBS-114A (REV. 2-8C)

U.S. DEPT. OF COMM. BIBLIOGRAPHIC DATA SHEET (See instructions)	1. PUBLICATION OR REPORT NO. NBSIR 80 - 2190	2. Performing Organ. Report No.	3. Publication Date

4. TITLE AND SUBTITLE

Characterizing the Interfiber Bonding of Currency Paper Pulps

5. AUTHOR(S)

Jack C. Smith and Elizabeth E. Toth

6. PERFORMING ORGANIZATION (If joint or other than NBS, see instructions)

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

7. Contract/Grant No.

8. Type of Report & Period Covered

Annual
10/1/79 thru 9/30/80

9. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS (Street, City, State, ZIP)

Bureau of Engraving and Printing
Treasury Department
Washington, D.C. 20234

10. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

A beaten currency stock was fractionated by fiber length. Handsheets of 2.5 g/m^2 mass per unit length were made from the unfractionated stock and the first three fractions. Similar handsheets were made from beaten and unbeaten kraft woodpulps. Scanning electron micrographs of these handsheets showed that currency pulp fibers attached to each other through many fine surface fibrils, and were held together by hydrophilic fines that acted as a glue. Woodpulp fibers were not fibrillated by beating nor broken into fines, but bonded by forming intimate contacts between their ribbon-like structures.

Specimens from the handsheets were extended in a tensile tester. Force-elongation behaviors were consistent with the behaviors expected from their morphologies. It was found that the force-elongation curves could be fit by a simple mathematical expression involving three parameters. These parameters are expected to prove useful in characterizing the bonding forces between pulp fibers in a paper.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

Adhesion of paper fibers; bonding of paper fibers; paper; paper fibers, bonding; paper pulps, characterization; paper, tensile tests.

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14. NO. OF PRINTED PAGES

15. Price